Anomalous thermopower and Nernst effect in $CeCoIn_5$: entropy-current loss in precursor state

Y. Onose[1], Lu Li[1], C. Petrovic[2] and N. P. Ong[1]
[1] Department of Physics, Princeton University, Princeton, NJ 08544, USA
[2] Department of Physics, Brookhaven National Laboratory, Upton, N.Y. 11973, USA

(Dated: February 1, 2008)

The heavy-electron superconductor $CeCoIn_5$ exhibits a puzzling precursor state above its superconducting critical temperature at $T_c = 2.3$ K. The thermopower and Nernst signal are anomalous. Below 15 K, the entropy current of the electrons undergoes a steep decrease reaching ~ 0 at T_c. Concurrently, the off-diagonal thermoelectric current α_{xy} is enhanced. The delicate sensitivity of the zero-entropy state to field implies phase coherence over large distances. The prominent anomalies in the thermoelectric current contrast with the relatively weak effects in the resistivity and magnetization.

PACS numbers: 71.27.+a,72.15.Eb,72.20.My,74.70.Tx

Several novel, interesting phenomena have been discovered in the anisotropic, heavy-electron superconductor $CeCoIn_5$ which has a critical transition temperature $T_c = 2.3$ K [1]. Evidence for d-wave pairing symmetry are seen in a number of experiments [2, 3, 4, 5]. The Fulde-Ferrell-Larkin-Ovchinikov (FFLO) state has been shown to exist below a temperature $T \sim 0.3$ K [6]. In a magnetic field $\mathbf{H}||\mathbf{c}$ (the c-axis), a crossover from non-Fermi liquid to Fermi liquid behavior has been reported [7, 8]. Magnetization results [10] reveal that a weak, unusual magnetic order begins to appear $\sim$20 K above T_c. This precursor state is mysterious and often associated with a "hidden" order parameter. Recently, Bel *et al.* [11] reported that a giant Nernst signal appears at $\sim$25 K. They reported that the sign of the Nernst signal is opposite to that expected from the vortex-Nernst signal, which has been intensively studied in cuprates [12, 13, 14].

Using thermopower, Nernst-effect and other transport measurements on high-purity $CeCoIn_5$, we have determined the full thermoelectric (Peltier) conductivity tensor $\boldsymbol{\alpha}$. In the precursor state at $H = 0$, a pronounced reduction of the thermopower reveals a remarkably steep loss of carrier entropy current. A weak field suppresses this low-entropy state. After subtraction of the thermal-Hall contribution to the Nernst signal and correcting its sign, we show that the enhanced Nernst signal correlates with the thermopower anomalies in the precursor state.

An applied temperature gradient $-\nabla T$ drives the charge current density $\mathbf{J} = \boldsymbol{\alpha}\cdot(-\nabla T)$. Because the total current density in the sample is zero, an internal electric field $\mathbf{E}$ arises to drive a counter current $\mathbf{J}' = \boldsymbol{\sigma}\cdot\mathbf{E}$, where $\boldsymbol{\sigma}$ is the conductivity tensor. The x and y components of $\mathbf{E}$ are observed, respectively, as the thermopower $S = E_x/|\nabla T|$ and the Nernst signal $e_N = E_y/|\nabla T|$ (we choose axes with $-\nabla T||\hat{\mathbf{x}}$ and $\mathbf{H}||\hat{\mathbf{z}}$). The thermopower, which involves balancing 2 counter charge currents $||\hat{\mathbf{x}}$, is just the ratio of 2 transport quantities, i.e. $S = \alpha/\sigma$, with $\alpha \equiv \alpha_{xx}$ and $\sigma \equiv \sigma_{xx}$ (we drop subscripts on diagonal quantities). However, the Nernst signal e_N is more involved.

In the simplest situation, the transverse gradient $-\partial_y T$ is negligible (isothermal case). Along $\hat{\mathbf{y}}$, we have the 2 off-diagonal currents $\alpha_{yx}(-\partial_x T)$ and $\sigma_{yx}E_x$, which must be cancelled by σE_y. In terms of the resistivity tensor $\boldsymbol{\rho} = \boldsymbol{\sigma}^{-1}$, we have [13]

$$e_N = \rho\alpha_{xy} + \rho_{xy}\alpha. \quad (1)$$

The Nernst signal e_N senses the sum of the off-diagonal Peltier current and the ordinary Hall current multiplied by S.

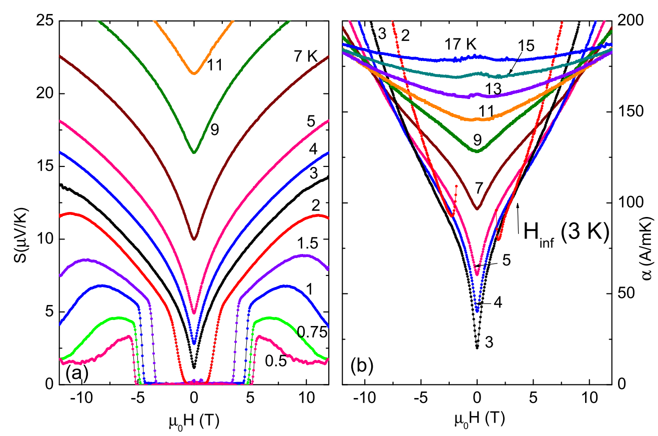

FIG. 1: (Panel a) The field dependence of the thermopower $S(H)$ of $CeCoIn_5$ at selected T ($\mathbf{H}||\mathbf{c}$). The zero-field anomaly deepens rapidly as T fall towards $T_c = 2.3$ K. Below T_c, $S(H)$ rises nearly vertically at H_{c2}, but continues to increase to a broad maximum (at $\sim$8 T at 1 K). Beyond the peak S decreases to a plateau value. Panel (b) shows curves of α vs. H from 2 to 17 K. As $T \to T_c$, α decreases (increases) if $|H|$ is smaller (larger) than H_{inf} (arrow drawn for 3 K).

However, when the thermal Hall conductivity κ_{xy} is large (as in $CeCoIn_5$ below 20 K [8]), a sizeable transverse gradient $-\partial_y T$ appears, which drives a charge current $||\hat{\mathbf{y}}$ via α (this is just the ordinary thermopower responding

to $-\partial_y T$). Instead of Eq. 1, we have [13]

$$e_N = \rho\alpha_{xy} - S\left[\frac{\sigma_{xy}}{\sigma} + \frac{\kappa_{xy}}{\kappa_e}\right], \qquad (2)$$

which expresses balancing the 3 off-diagonal currents $\sim \alpha_{xy}$, $\sim \sigma_{xy}$ and $\sim \kappa_{xy}$ by the current σE_y. Thus to obtain α_{xy} from e_N, we should subtract the charge and thermal Hall currents. (We adopt the sign convention [13, 14] that e_N shares the sign of α_{xy}, i.e. when $\alpha_{xy} > 0$ and dominant, e_N is positive. Vortex flow in a superconductor gives a positive e_N.)

We first describe the field dependence of S and the inferred α. At low T, S displays a very interesting field dependence (Fig. 1a). Above ∼15 K, S is nearly insensitive to H aside from a weak cusp-like anomaly at H= 0. With decreasing T, the anomaly deepens and imparts a strong H dependence that extends to ∼12 T. In the interval 3-7 K, the zero-H cusp is bracketed by a steep H-linear dependence (at 3 K, S increases 14-fold between $H = 0$ and 12 T). Below T_c, S rises nearly vertically from zero at H_{c2} (the upper critical field), attains a broad maximum (8.1 T at 1 K), and settles to a plateau value at large H.

Using the magnetoresistance ρ vs. H from Ref. [8], we have converted the S-H curves into α vs. H (Fig. 1b). At 17 K, α displays the weak H^2 dependence characteristic of the normal-state thermoelectric conductivity α^n given by the Mott expression $\alpha^n = \frac{\pi^2}{3}\frac{k_B^2 T}{e}\left[\frac{\partial\sigma}{\partial\epsilon}\right]_\mu$, where k_B is Boltzmann's constant, e the elemental charge, ϵ the energy and μ the Fermi level.

However, as $T \to$3 K, the minima in α deepen to cusps even sharper than those in S. Below 15 K, the curve of α displays an inflexion point at a field H_{inf} where $\partial^2\alpha/\partial H^2 = 0$ (H_{inf} increases from 3.7 T at 3 K to 9.4 T at 9 K). The inflexion field reveals 2 factors that pull α in opposite directions. As T decreases, α increases if $|H| > H_{inf}$, but decreases for $|H| < H_{inf}$. The cusp anomaly is dominant for $|H| < H_{inf}$. Further, the field scale defining the cusp sharpness (∼100 G at 3 K) suggests that the field is spoiling the phase coherence of the wave function at low T (see below).

A notable feature is that the anomaly in α is much larger than that in ρ. For e.g., in the interval $H = 0 \to H_{inf}$ defining the anomaly, α increases more than five-fold at 3 K, whereas the cusp in ρ constitutes only 20% of the total resistivity [8]. The unusually large cusps in S reflects this huge discrepancy. We return to these features after describing the Nernst results.

In Ref. [11], an enhanced Nernst signal was reported below ∼25 K, but with a sign opposite to that of the vortex-Nernst effect [12, 13, 14]. Our measurements of e_N (Fig. 2) are nominally consistent in magnitude with Ref. [11]. However, we find that the sign of e_N is positive (after some correspondence, the authors of Ref. [11] have confirmed that the sign is positive [9]).

Using S and the tensors $\boldsymbol{\sigma}$ and $\boldsymbol{\kappa}$ measured in Ref. [8], we now use Eq. 2 to extract α_{xy}. The separate

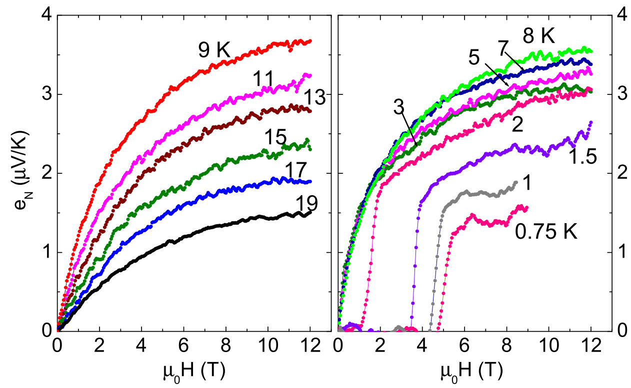

FIG. 2: The observed Nernst signal e_N vs. H in $CeCoIn_5$ from T = 9 to 19 K (Panel a) and from 0.75 to 8 K (b). Below T_c, e_N undergoes a sharp jump from zero at H_{c2}. Throughout, e_N includes a significant contribution from a current driven by κ_{xy}. The sign of the Nernst effect is positive.

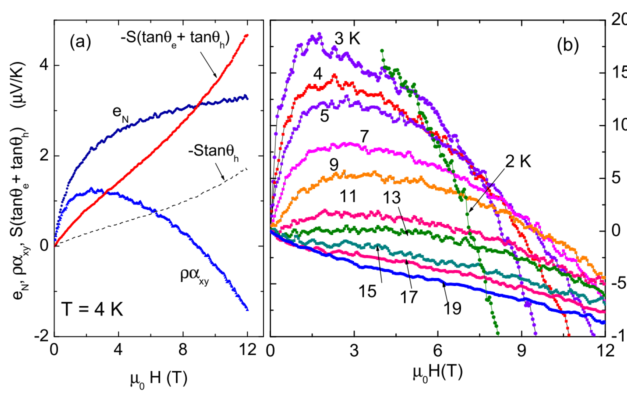

FIG. 3: (Panel a) Separation of the observed Nernst curve e_N at 4 K into the off-diagonal Peltier term $\rho\alpha_{xy}$ and the charge-and-thermal Hall term $-S(\frac{\sigma_{xy}}{\sigma} + \frac{\kappa_{xy}}{\kappa})$. As shown by the dashed curve, 30-40 % of the latter derives from κ_{xy}. $S > 0$ (hole-like), while $\sigma_{xy} < 0$ and $\kappa_{xy} < 0$ (electron-like). (Panel b) The curves of α_{xy} vs. H derived from e_N using Eq. 2. At 17 and 19 K, α_{xy} is nearly H-linear, consistent with a qp origin. The plotted quantity is the sum of 2 terms (Eq. 3). The broadly peaked profile at lower T (<15 K) is the anomalous term α^s_{xy}. At the lowest T, the negative qp term α^n_{xy} pulls α_{xy} to large negative values (-100 A/mK at 12 T and 2 K).

contributions are shown in Fig. 3a at T = 4 K. The curve of e_N is the sum of $\rho\alpha_{xy}$ and the augmented Hall term $-S(\frac{\sigma_{xy}}{\sigma} + \frac{\kappa_{xy}}{\kappa})$. We note that, in $CeCoIn_5$, $S > 0$ (hole-like), while $\sigma_{xy} < 0$ and $\kappa_{xy} < 0$ (electron-like). Roughly $\frac{1}{3}$ of the Hall term derives from the current due to κ_{xy} (dashed line).

The derived field profiles of α_{xy} are highly instructive

(Fig. 3b). At the high-T end (17 and 19 K), the curves are H-linear, consistent with the off-diagonal thermoelectric response of quasi-particles in moderate fields. We identify this as the qp background term, atop of which a positive contribution to α_{xy} emerges as $T \to T_c$. Referring back to e_N in Fig. 2, we may now see that, at temperatures above 15 K, the "enhanced" Nernst signal with pronounced curvature merely reflects the large charge and thermal Hall currents σ_{xy} and κ_{xy}. These contribute to the Nernst signal, but are not intrinsic to the off-diagonal thermoelectric response.

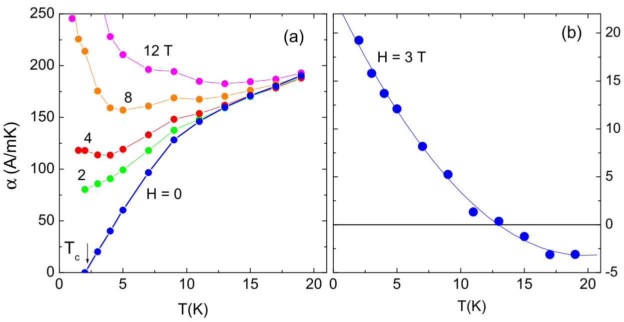

FIG. 4: (Panel a) The T dependence of α with H fixed at selected values. At $H = 0$, the curve of α decreases monotonically to zero at T_c. A weak H readily suppresses this downward trend. At large H (> 5 T), α is strongly enhanced because of steep increase of ℓ. Panel (b) shows the T dependence of α_{xy} with H fixed at 3 T. The increase in α_{xy} below 15 K mirrors the decrease in α in weak H in Panel (a). Above 15 K, α_{xy} approaches the negative qp term α^n_{xy}.

The pattern of the curves in Fig. 3b suggests that α_{xy} is comprised of 2 terms, viz.

$$\alpha_{xy} = \alpha^n_{xy} + \alpha^s_{xy}. \tag{3}$$

where α^n_{xy} is the negative qp term and α^s_{xy} the positive anomalous term. Below 15 K, the latter swells rapidly with a characteristic field profile that peaks at relatively low fields (2-3 T) and then decays slowly at large H. The T dependence of α_{xy} at a fixed H (3 T) is shown in Fig. 4a. It is apparent that the anomalous term appears as a positive contribution on top of a negative, H-linear background.

In Fig. 3b, the curves below 5 K exhibit a steep decrease to large negative values at high fields. As reported in Ref. [8], measurements of κ_{xy} and σ_{xy} reveal that, below 6 K, the electronic mean-free-path ℓ is sharply enhanced in large H because of field-suppression of scattering by spin disorder [8]. We identify the high-field changes in α_{xy} with strong enhancement of the qp term α^n_{xy} which scales like ℓ^2 (α^n_{xy} satisfies the Mott expression as α^n, with σ replaced by σ_{xy}). The increase in ℓ strongly enhances α^n_{xy}, which is intrinsically negative, as we noted at 17-19 K. At 2 K, the qp contribution is so large that it pulls α_{xy} to very large negative values (-100 A/mK at 12 T). Hence, at high fields, the total off-diagonal term α_{xy} is dominated by the steep growth of the negative qp term, but at low fields, it is dominated by the positive anomalous term. This is just the 2 disparate trends separated by H_{inf} in the curves of α (Fig. 1b), but now observed in the off-diagonal channel.

We now return to the implications of the thermopower. Figure 4a shows the T dependence of $\alpha(T, H)$ at several fixed H. Starting at 20 K, the zero-field curve $\alpha(T, 0)$ initially decreases with a modest slope, but below 15 K, it accelerates to fall steeply to zero close to T_c.

There are 2 unusual aspects of $\alpha(T, H)$. First, in conventional superconductors, α (and S) display a step-like decrease to zero at T_c, whereas α here is already fully suppressed just above T_c. By Onsager reciprocity, $\alpha = \tilde{\alpha}/T$, where $\tilde{\alpha}$ defines the heat current $\mathbf{J}_h$ produced by $\mathbf{E}$ (with $\nabla T = 0$), viz. $J_h = \tilde{\alpha}E$, so that we may regard $J_S = \alpha E$ as the entropy current density generated by $\mathbf{E}$ (whence S is the entropy transported per unit charge). In this view, the sharp decrease of $\alpha(T, 0)$ in the broad 13-K interval above T_c (Fig. 4a) implies a dramatic loss of entropy carried by the conduction electrons. The loss in carrier entropy is a new feature of the precursor state above T_c in $CeCoIn_5$.

Secondly, as shown in Fig. 1b, the zero-field behavior is very field sensitive. At 3 K, the cusp is rounded in a very weak field (100 G) and nearly completely suppressed at H_{inf} (3.7 T). A field of 100 G is equivalent to a magnetic length $\sqrt{\hbar/eB} \sim$ 2,600 Å. It is unlikely that field sensitivity on such long lengths can be explained semi-classically. Instead, it is indicative of field decoherence of an electronic wave function that retains phase coherence over large distances. Two examples displaying such weak-field sensitivity are the suppression of weak Anderson localization, via dephasing of paths related by time-reversal symmetry, and field suppression of phase coherence in the fluctuation regime of superconductors.

Within our accuracy, the onset temperature of α^s_{xy} is identical with that of the cusp anomaly in α (Fig. 1b). The actual anomalous contribution detected in the Nernst signal shares the same origin as the anomalous term in the thermopower; both appear below 15 K rather than 25 K. An important feature, however, is that α is much more field sensitive than α^s_{xy}, as may be seen by comparing Figs. 1b and 3b. The latter survives to fields larger than 6 T whereas the cusp in α is affected by very weak H.

In analogy with the cuprates [13, 14], an appealing candidate for the precursor state above T_c would seem to be the vortex-liquid scenario in which the Cooper-pair condensate lacks phase rigidity on long length scales because of spontaneous (anti)vortices. The scenario accounts for the steep fall of α as $T \to T_c$ because regions of the sample in which the condensate retains phase coherence have reduced entropy. In a field, vortices inserted by H lead to rapid phase decoherence of these regions to produce the steep increase observed in α. Moreover, the flow of vortices in the applied gradient generates a large, posi-

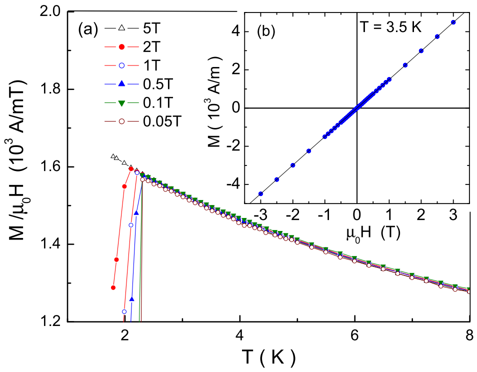

FIG. 5: (a) The magnetization M vs. T above T_c in $CeCoIn_5$ expressed as a susceptibility $M/\mu_0 H$. Curves measured by SQUID magnetometry at fields $\mathbf{H}||\mathbf{c}$ from 0.05 T to 5 T coincide. Panel (b) shows the strictly H-linear variation of M at 3.5 K. The susceptibility χ equals $+1.879\times10^{-3}$. Within our resolution, M is linear in H; an H-dependent, diamagnetic contribution is not detected.

tive Nernst signal as observed. Insofar as the anomalous term α^s_{xy} comes from phase-slip events arising from the passage of individual vortices, the Nernst signal does rely on having regions with long-range phase coherence, so it survives to large fields. This scenario is compatible with the rapid changes in α in weak H, as well as the relative robustness of α_{xy} to moderately large fields.

The vortex liquid above T_c should exhibit a sizeable diamagnetic signal, as has been confirmed in the case of cuprates [15]. However, our measurements of magnetization M have not uncovered such diamagnetism in $CeCoIn_5$ (Fig. 5a). Above T_c, the magnetization is dominated by a T-dependent, paramagnetic susceptibilty that is large ($\chi\sim10^{-3}$) [10]. Within our resolution, M is observed to be strictly linear in H from 0 to 5 T at 3.5 K, with no trace of a diagmagnetic fluctuation contribution (Fig. 5b). Moreover, below T_c, the upper critical field H_{c2} is sharply defined and given by the mean-field form $H_{c2}\sim(T_c-T)$, very unlike the situation in cuprates. Clarification of the magnetization may require experiments that can resolve a fluctuating M at the level of a 1-10 A/m [14].

The thermoelectric current response in a field has revealed several unusual characteristics of the precursor state in $CeCoIn_5$ which appear below 15 K. In zero H, the striking decrease of S and α towards $\sim$0 near T_c implies a sharp loss of the entropy current carried by quasiparticles. The striking field sensitivity of α implies that the zero-entropy feature occurs in regions with long-range phase coherence. Concurrently, the off-diagonal current $\alpha_{yx}(-\nabla T)$ gains an anomalous positive contribution, which is readily distinguised from the negative qp contribution. In contrast to these very prominent anomalies in the thermoelectric current, the modification to the resistivity is quite modest [8]. The contribution to the magnetization is currently below our resolution.

We acknowledge valuable disussions with K. Behnia, Y. Matsuda, C. Broholm and L. Taillefer. Y. O. thanks the Nishina Memorial Foundation for partial support. Research at Princeton University and the Brookhaven National Laboratory was supported by the U.S. National Science Foundation (DMR 0213706) and the U.S. Department of Energy (DE-Ac02-98CH10886), respectively.

[1] C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarro, J. D. Thompson, Z. Fisk, and P. Monthoux, J. Phys. Cond. Matter **13,** L337 (2001).
[2] R. Movshovich, M. Jaime, J. D. Thompson, C. Petrovic, Z. Fisk, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **86,** 5152 (2000).
[3] K. Izawa, H. Yamaguchi, Y. Matsuda, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. Lett. **87**, 057002 (2001).
[4] R. J. Ormeno *et al.*, Phys. Rev. Lett. **88**, 047005 (2002).
[5] H. Aoki *et al.*, J. Phys. Cond. Matter **16**, L13 (2004).
[6] H. A. Radovan *et al.*, Nature (London) **425,** 51 (2003); A. Bianchi *et al.,* Phys. Rev. Lett. **91,** 187004 (2003); K. Kakuyanagi *et al.,* **94,** 047602 (2005).
[7] J. Paglione *et al.*, Phys. Rev. Lett. **91**, 246405 (2003); A. Bianchi et al., Phys. Rev. Lett. **91,** 257001 (2003).
[8] Y. Onose, N. P. Ong, and C. Petrovic, *preprint* 2006.
[9] K. Izawa *et al.*, cond-mat/0704.1970.
[10] T.P. Murphy *et al.*, Phys. Rev. B 65, 100514 (2002); T. Tayama *et al.*, Phys. Rev. B 65, 180504 (2002).
[11] R. Bel *et al.*, Phys. Rev. Lett. **92**, 217002 (2004).
[12] Z. A. Xu, N. P. Ong, Y. Wang, T. Kakeshita, and S. Uchida, Nature (London) **406,** 486 (2000).
[13] Y. Wang, S. Ono, Y. Onose, G. Gu, Y. Ando, Y. Tokura, S. Uchida, and N. P. Ong, Science **299,** 86 (2003).
[14] Y. Wang, Lu Li, and N. P. Ong, Phys. Rev. B **73**, 024510 (2006).
[15] Y. Wang *et al.*, Phys. Rev. Lett. **95,** 247002 (2005); L. Li *et al.*, Europhys. Lett. **72**, 451 (2005).